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Effects of Humidity Swings on Adsorption Columns for Air Revitalization: Modeling and Experiments

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Abstract

Air purification systems are necessary to provide clean air in the closed environments aboard spacecraft. Trace contaminants are removed using adsorption. One major factor concerning the removal of trace contaminants is relative humidity. Water can reduce adsorption capacity and, due to constant fluctuations, its presence is difficult to incorporate into adsorption column designs.

This report describes progress made during the first year of a two-year Joint Research Interchange between the NASA-Ames Research Center and the University of Virginia. The goal of this research is to allow for better design techniques in trace contaminant adsorption systems, especially for feeds with water present. Experiments and mathematical modeling research on effects of humidity swings on adsorption columns for air revitalization is being carried out by Mr. W. Scot Appel, a graduate student in the Department of Chemical Engineering at the University of Virginia, under the joint active direction of Prof. M. Douglas LeVan of the University of Virginia and Dr. John E. Finn of the NASA-Ames Research Center.

Progress was made on both the experimental and modeling aspects of the problem during this first year. Mr. Appel spent all of June and July, 1994 at the NASA-Ames Research Center, Moffett Field conducting experimental work with a fixed-bed adsorption column. The remainder of his research effort was spent at the University of Virginia writing computer code for the model and studying relevant literature.

Introduction

The continued survival of human life in a closed environment requires a breathable air supply. In the limited volumes found in such places as submarines, aircraft, and spacecraft, an air revitalization system is essential to meet this demand. To insure human comfort and safety, large concentrations of carbon dioxide as well as contaminants which are present in trace amounts must be removed from the air. The elimination of trace contaminants, particularly components of high volatility, can be the more difficult problem as the low concentrations involved provide a small driving force for separation.

For NASA, removal of trace contaminants is not a trivial problem. The atmosphere of a spacecraft is constantly corrupted from both the off-gassing of equipment as well as the metabolic products of the crew. Many contaminants pose health hazards to the crew members, even at very low concentrations. Some trace contaminants which are of primary concern are chlorofluorocarbons, benzene, MEK, low molecular weight alcohols, and xylenes as well as metabolic products such ammonia, carbon monoxide, methane, and indole [Finn, 1993]. The removal or these and more than 200 other contaminants found during space missions presents NASA with a difficult challenge [Leban & Wagner, 1989].

Based on a model of contaminant loadings, a trace contaminant control system was developed at Lockheed Missiles and Space Company for use in the proposed Space Station [Lamparter, 1989]. The purification system includes an acid-impregnated activated carbon adsorption bed. The acid removes ammonia and the activated carbon adsorbs many of the organic pollutants. Following the adsorption bed is a catalytic oxidizer which will destroy the light hydrocarbons, such as methane, that do not adsorb well onto activated carbon, but would be quickly poisoned by any heavier hydrocarbons which manage to escape from the activated carbon bed. Finally, the flow will pass through a LiOH bed to remove any acid gases created in the oxidizer.

Although this purification system sounds adequate, problems may arise. The adsorption bed is designed to operate for 90 days before it is replaced. This 90 day cycle is based on a few, design-driving, contaminants which are expected to breakthrough first. The actual performance of the adsorption bed in the presence of the over 200 pollutants cannot be accurately known. Interactions between species may cause unexpected adsorption behavior. Also, the presence of water (humidity) in the inlet stream to adsorption bed can affect contaminant breakthrough times. By design, trace contaminants enter the bed at concentrations near 0.002 mol/m3; however, water concentration can vary from about 0.15 to 0.45 mol/m3 (25% RH to 75% RH). Variations of this magnitude are difficult to account for in adsorption column design [Space Station Architectural Control Document, 1991].

The effect that a 50% variation in RH will have on contaminant breakthrough is not known for sure; however, some possibilities can be envisioned based on knowledge of the coadsorption of water and hydrocarbons [Rudisill et al., 1992; Eissmann and LeVan, 1993]. Water has been shown to reduce the adsorption capacity of an activated carbon bed towards some contaminants [Finn, 1993]. Water, acting as an adsorbing species, can compete with contaminants for adsorption space. Also, a humidity swing, represented by a pulse through the column, can displace adsorbed species which are either less strongly adsorbed than water or are hydrophobic [LeVan & Finn, 1993]. One additional result of premature breakthrough is poisoning of the catalytic adsorption bed. On long term space missions, it is not feasible to replace equipment more frequently than allowed by design. Premature breakthrough is simply not acceptable.

Current adsorption bed models utilized by NASA neither account for the interactions between species nor treat water as an absorbable species. Humidity is accounted for simply by increasing the size of the activated carbon bed. While this solution works for short missions, it will not be practical for long term space travel where size and efficiency are more important. Also, should a humidity pulse occur, due to crew cycles or equipment changes, it is highly desirable to know from predictions the effect at the bed outlet and to have made allowances in design based on those predictions.

The goal of this research is to develop a dynamic model which can predict the effect of humidity swings on activated carbon adsorption beds used to remove trace contaminants from the atmosphere in spacecraft. Specifically, the model will be incorporated into a computer simulation to predict contaminant concentrations exiting the bed as a function of time after a humidity swing occurs. Predicted breakthrough curves will be compared to experimentally measured results.

The two major aspects of this research are a mathematical model and an experimental apparatus. Once the model and simulation code have been used to generate breakthrough curves, data will be collected with the apparatus to validate and improve the predictions. During the past year, work has been done on both the model and the apparatus.

Model

A mathematical model was developed from differential mass and energy balances on the adsorption bed and is similar to the one used of Davis & LeVan [1989]. The model is be based on an inlet flow containing three contaminants (ethanol, CFC-113, and water) carried in an inert flow of nitrogen. Ethanol and CFC-113 were chosen based on their importance to design as well as their respective hydrophilic and hydrophobic natures, which is important during adsorption. The complete model includes 7 coupled differential equations: a material balance and a rate equation for each of the three absorbable components and an energy balance.

Picture a well mixed gas stream entering an adsorption column. The gas contains 3 absorbable species [(i) where $i = 1,2,3$] carried in an inert flow of nitrogen. Initially, the species are present at their respective inlet concentrations. As soon as the flow enters the bed, these concentrations change. Variations in concentration are caused by adsorption and axial dispersion. Due to the low contaminant concentrations as well as the nature of the problem, no axial dispersion term has been included in the model. Future revisions may include dispersion terms so their effect, if any, can be observed.

Neglecting axial dispersion, a material balance can be made on each component. Mathematically, this is represented by

$$\rho_b \frac{\partial q_i}{\partial t} + \varepsilon' \frac{\partial c_i}{\partial t} + \varepsilon \frac{\partial (v c_i)}{\partial z} = 0$$

where q is adsorbed-phase concentration, c is gas-phase concentration, ε' is to total bed voidage, ε is the packing void fraction, and v is interstitial gas velocity. The first and second terms represent the accumulation of (i) in the absorbed phase and gas phase respectively.

With the material balance written in this form, the energy equation follows naturally. The internal energy of a molecule is greater in the gas phase than the adsorbed phase since it has freedom to move as a gas. This difference in related to the heat of adsorption. Heat is released when a species adsorbs, causing the bed temperature to rise. Therefore, a temperature gradient will exists in the bed. Also, there will exist heat losses from the bed to the surroundings, necessitating the use of a heat transfer term through the bed wall.

The mathematical statement of the energy balance is very similar to the material balance. Energy accumulates as internal energy in the adsorbed and fluid phases and flows in the form of fluid enthalpy. The general form is

$$\rho_b \frac{\partial u_s}{\partial t} + \varepsilon' \frac{\partial u_f}{\partial t} + \varepsilon \frac{\partial (v h_f)}{\partial z} + \frac{2U}{R_c}(T - T_{amb}) = 0$$

This equation is not in its most usable form. Substituting appropriate definitions for internal energies and enthalpies gives, after some algebraic manipulations

$$\rho_b[(cp_{bed} + (\Sigma_i q_i) cp_a)\frac{\partial T}{\partial t} - (\Sigma_i \lambda_i \frac{\partial q_i}{\partial t})] +$$

$$\varepsilon' [(\Sigma_i c_i)\frac{\partial T}{\partial t} - \frac{\partial P}{\partial t}] + \varepsilon v (\Sigma_i c_i) cp_{gas} \frac{\partial T}{\partial z} + \frac{2U}{R_c}(T - T_{amb}) = 0$$

Along with the material and energy balance equations, a rate equation for $\partial qi / \partial t$ is utilized. This term is approximated by a linear driving force of the form:

$$\frac{\partial q_i}{\partial t} = k_{qi}(q_i^* - q_i)$$

where k_{qi} is a mass transfer coefficient and q_i^* is the value of q_i in equilibrium with c_i.

The material and energy balances are partial differential equations in time and space. In the present form of the model, the spatial derivatives are approximated using backward finite differences. This reduces the equations to a large set of coupled ordinary differential equations in time. In effect, backward differences divides the column into a series of stages, or mixing cells as shown in Figure 1.

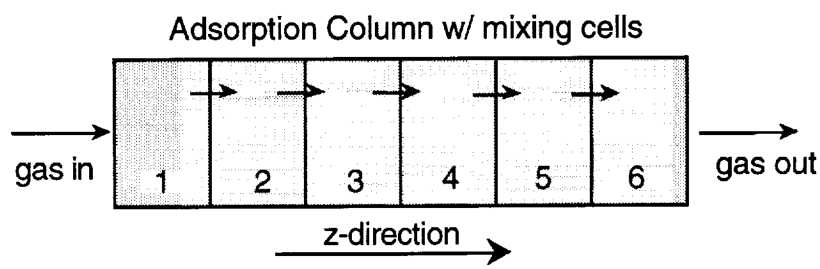

Figure 1: Fixed-bed as a cascade of mixing cells.

The equations are solved simultaneously for the entire adsorption column. We use the Gear's-method ordinary differential equation solver LSODE, the Livermore Solver for Ordinary Differential Equations. As part of the computational technique, the column is divided into a number of mixing cells. The greater the number of mixing cells, the more accurate the solution. Since each mixing cell is coupled with the others, the vector sent to LSODE requires all first derivatives with respect to time of each of the 7 unknowns in every mixing cell.

Perhaps the most difficult problem facing this research is the need for accurate equilibrium data. Without equilibrium isotherm data, it is difficult to correctly predict breakthrough curves. As part of other research, isotherm correlations for components of interest are currently being developed within Prof. LeVan's group at the University of Virginia. Currently, modified multicomponent Langmuir isotherms have been incorporated into the simulation package.

Apparatus

The experimental apparatus which will be used to test the validity of the model is located at NASA Ames Research Center. This apparatus, shown in Figure 2, allows the operator to manipulate the humidity level entering the bed by controlling the percentage of the inert stream (nitrogen) which is humidified. Initially, the inert stream is divided into 2 flows. One stream is then passed in cross flow with water where its relative humidity reaches nearly 100%. The inert streams (humid & dry) then recombine and mix with some trace contaminants. Changing the humidity is as easy as changing the set point on 2 mass flow controllers. This stream of known concentration and humidity enters the adsorption bed. Upon exiting the bed, compositions are determined using a gas chromatagraph (GCMS). All of the relevant of readings are sent to a computer via a data acquisition system for easy analysis.

The apparatus will allow for many types of experiments. Simulations can be made to model actual humidity fluctuations which may occur in the Space Station as well as controlled bed loading schemes with subsequent humidity changes which are more easily studied and repeated.

Due to the location of the apparatus, experimental work was conducted during June and July, 1994. The apparatus was assembled and brought into working order. An experimental program is planned beginning in May, 1995.

Future Plans

As indicated, the goal of this research is to aid NASA design teams in developing better trace contaminant control systems. There are many sets of experiments that will yield information relevant to the design of adsorption columns. Simulations which are made will be validated by experiments beginning in May, 1995.

Initial simulations will involve uniformly loading the adsorption column with ethanol and/or CFC-113 in the presence of low humidity. Step changes in humidity levels will then be made and the concentrations exiting the bed will be observed. These simulations will give an indication of the effects rapid humidity swings can have aboard the Space Station perhaps due to equipment malfunction. This experiment can be compared to one in which a uniformly loaded bed is subjected to a cycled humidity changes such as would arise from diurnal crew cycles.

The effectiveness of the adsorption bed can be studied at different humidity levels. This can be accomplished by using fixed inlet concentrations of the contaminants and a fixed value for relative humidity. Breakthrough curves can be compared for different relative humidities. This may help locate an optimal relative humidity that should be sought for the Space Station.

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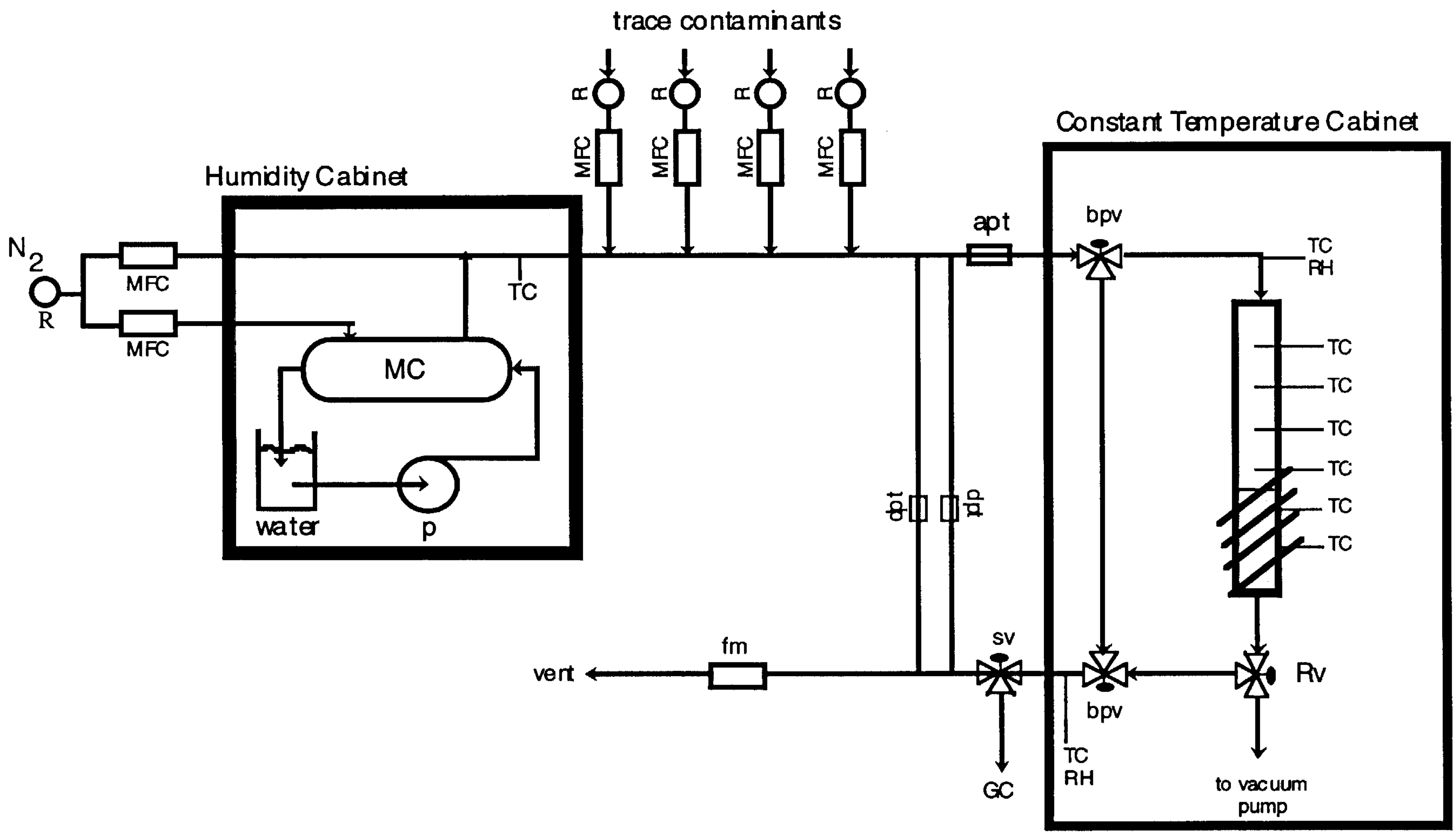
Figure 2: Experimental Apparatus



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